Supporting Information

Cobalt-based Nanosheet Arrays as Efficient Electrocatalysts for Overall Water Splitting

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Catalogue

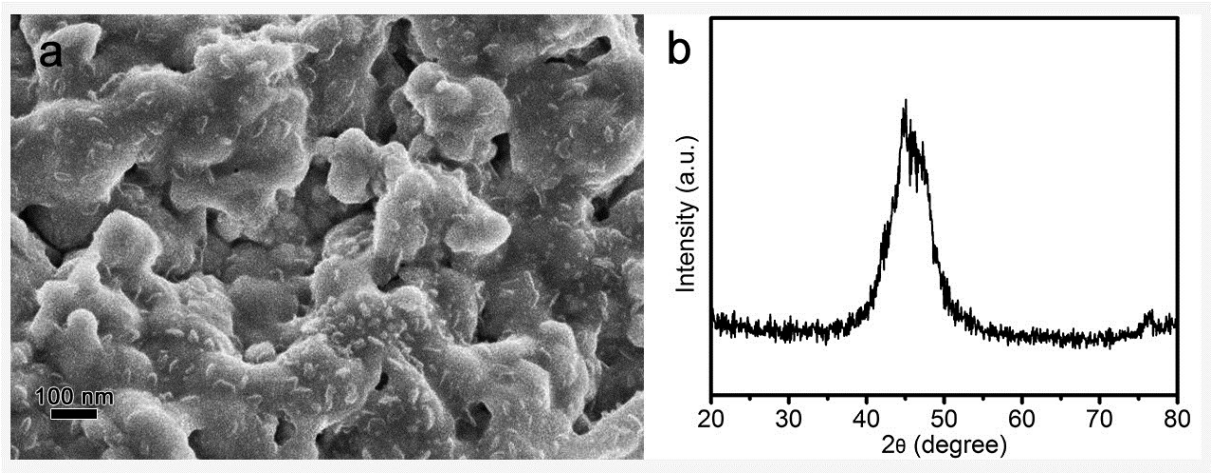

Figure S1. (a) SEM image and (b) XRD spectrum of as-deposited cobalt alloy.

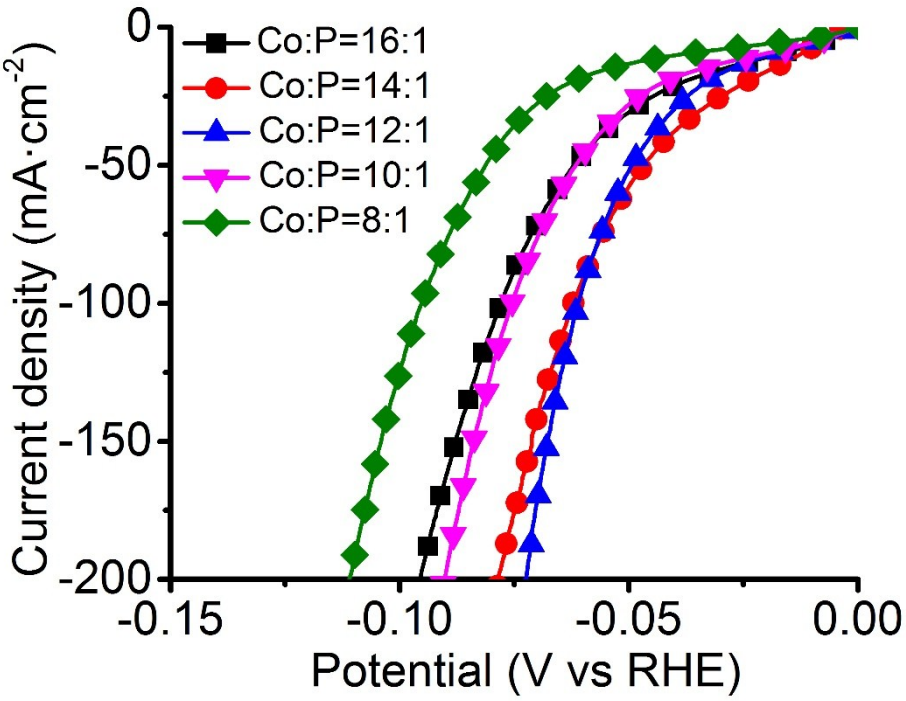

Figure S2. Linear sweep voltammetry of Co NS (for HER reaction) synthesized with different $CoSO_4$:NaH_2PO_2 molar ratio.

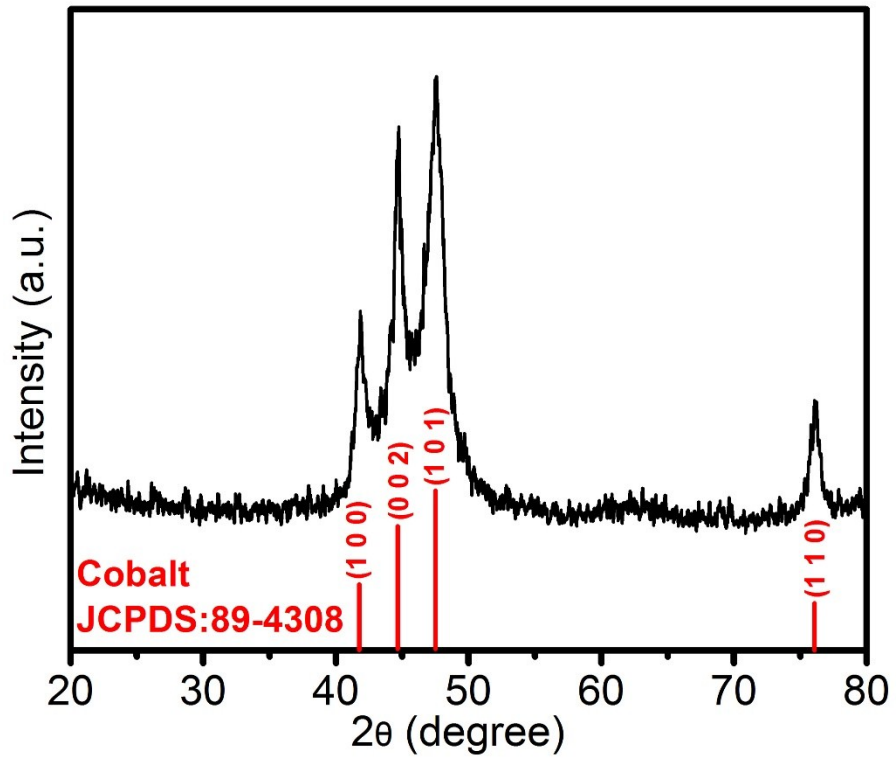

Figure S3. XRD spectrum of Co NS sample**.**

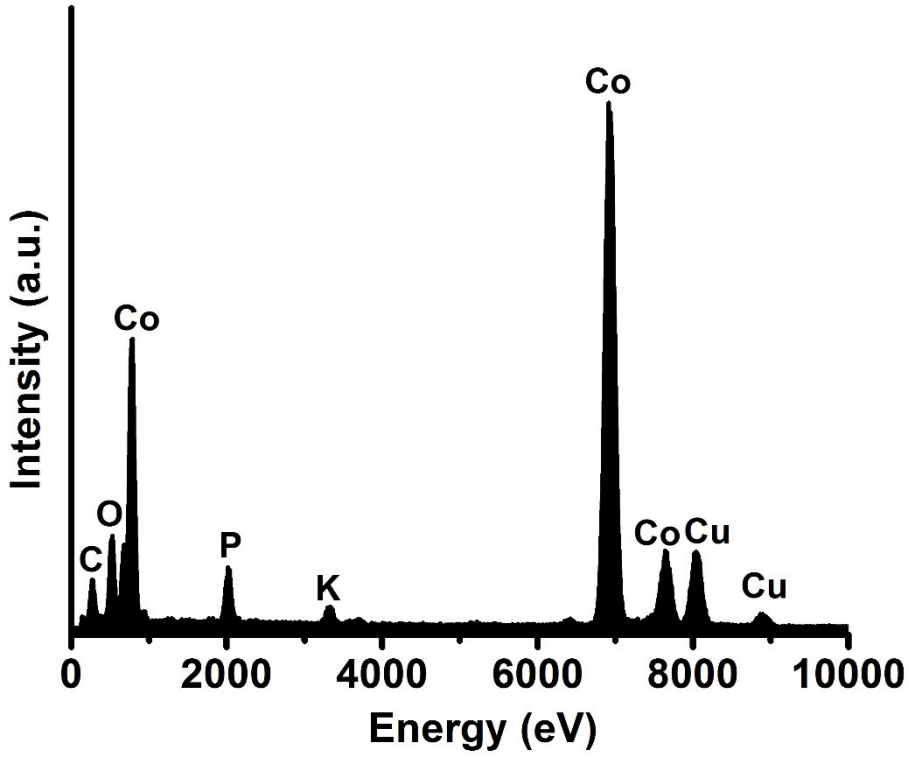

Figure S4. EDX spectrum of Co NS sample.

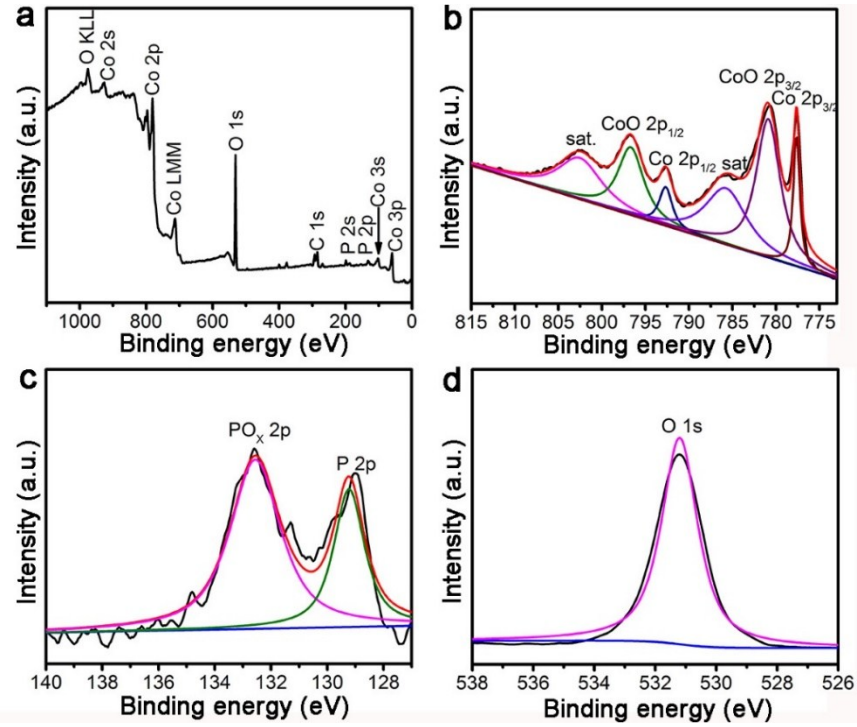

Figure S5. a) XPS survey spectrum for Co NS; core level of XPS spectrum in the (b) Co 2p, (c) P 2p and (d) O 1s for Co NS.

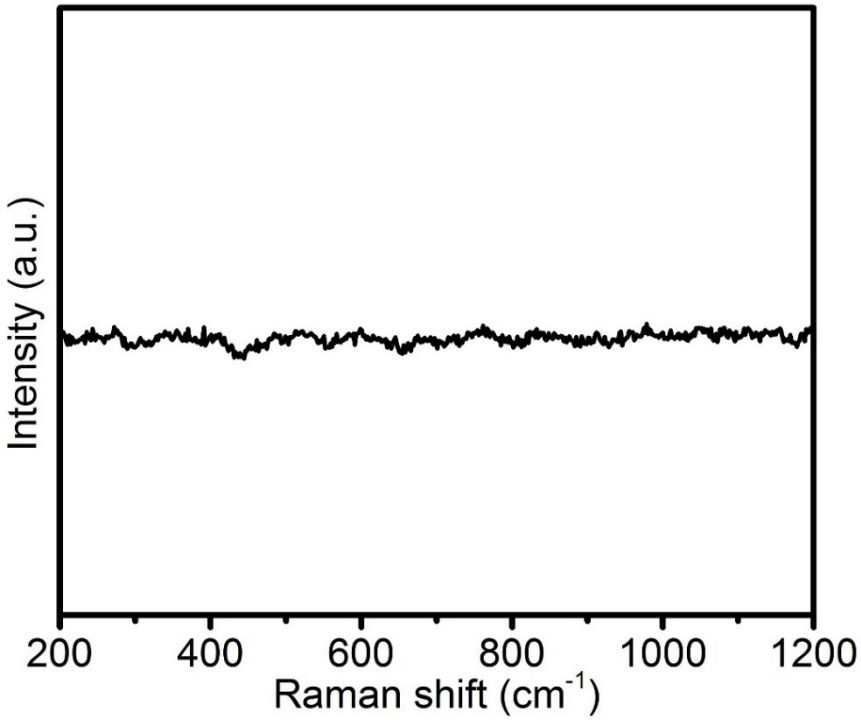

Figure S6. Raman spectrum of Co NS sample. There are no obviously peaks in the Raman spectrum of the Co NS before and after the 1h of pre-reduction, which further confirms that the surface of the Co NS catalysts is comprised of cobalt metal.

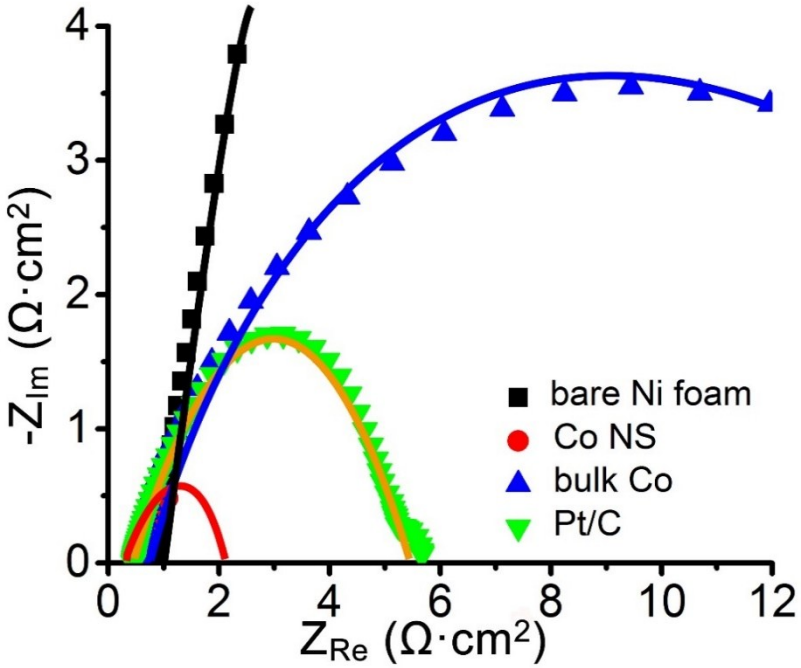

Figure S7. Nyquist plots of bare NF, Co NS, bulk Co and 20% Pt/C electrodes for HER in 1.0 M KOH.

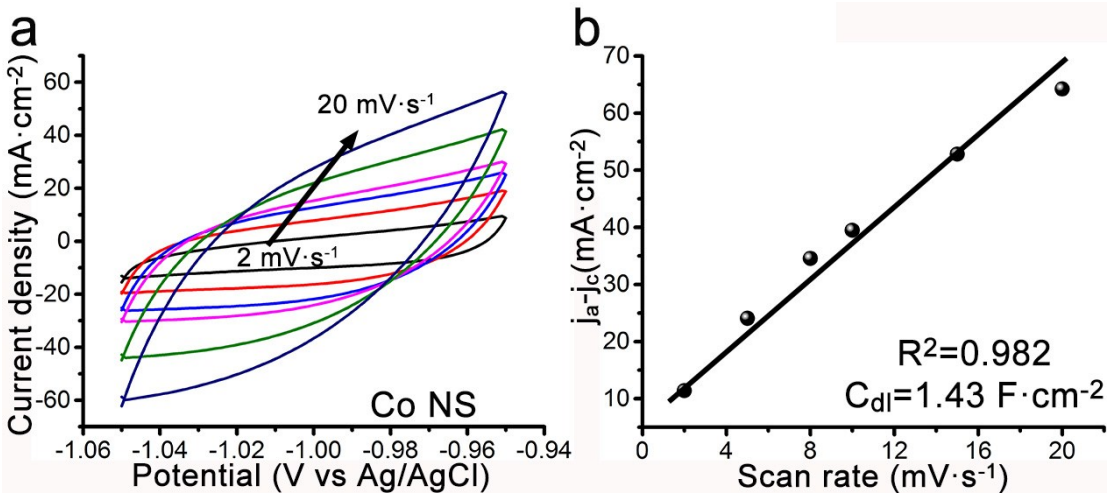

Figure S8. (a) Cyclic voltammograms recorded for the Co NS electrode at various scan rates in the non-Faradaic region in 1.0 M KOH. (b) Scan rate dependence of the current densities of the Co NS electrode at -1 V vs Ag/AgCl . The slope of the lines in (b) is the effective electrochemically active surface area of Co NS electrodes.

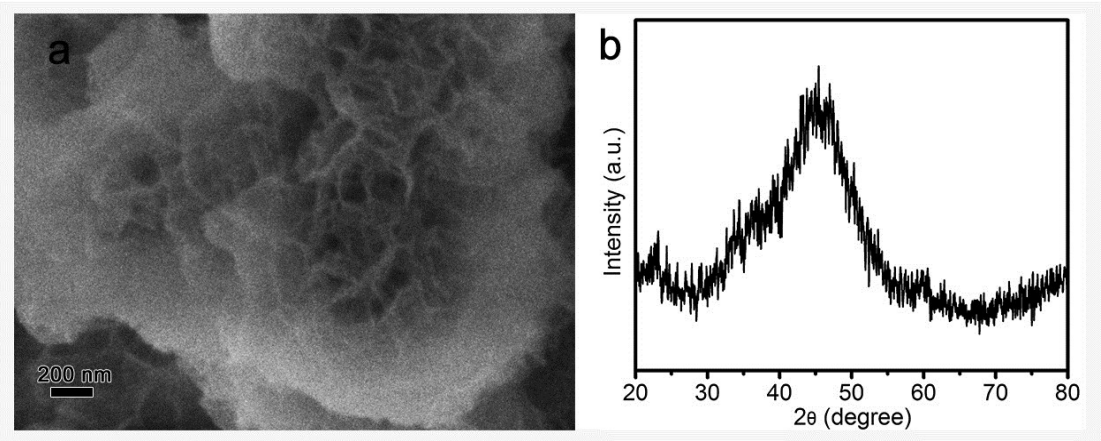

Figure S9. (a) SEM image and (b) XRD spectrum of as-prepared CoFeB alloy.

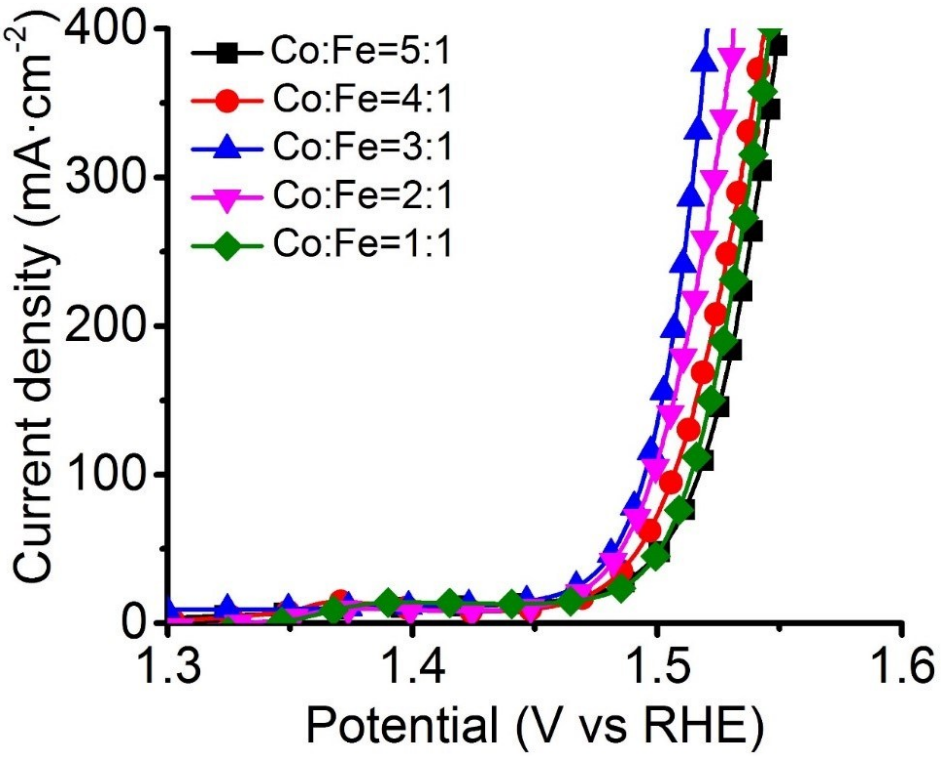

Figure S10. Linear sweep voltammetry of CoFeBO NS (for OER reaction) synthesized with different $CoCl_2$: $FeSO_4$ molar ratio. The molar ratio of B: (Co+Fe) was fixed at 4:1.

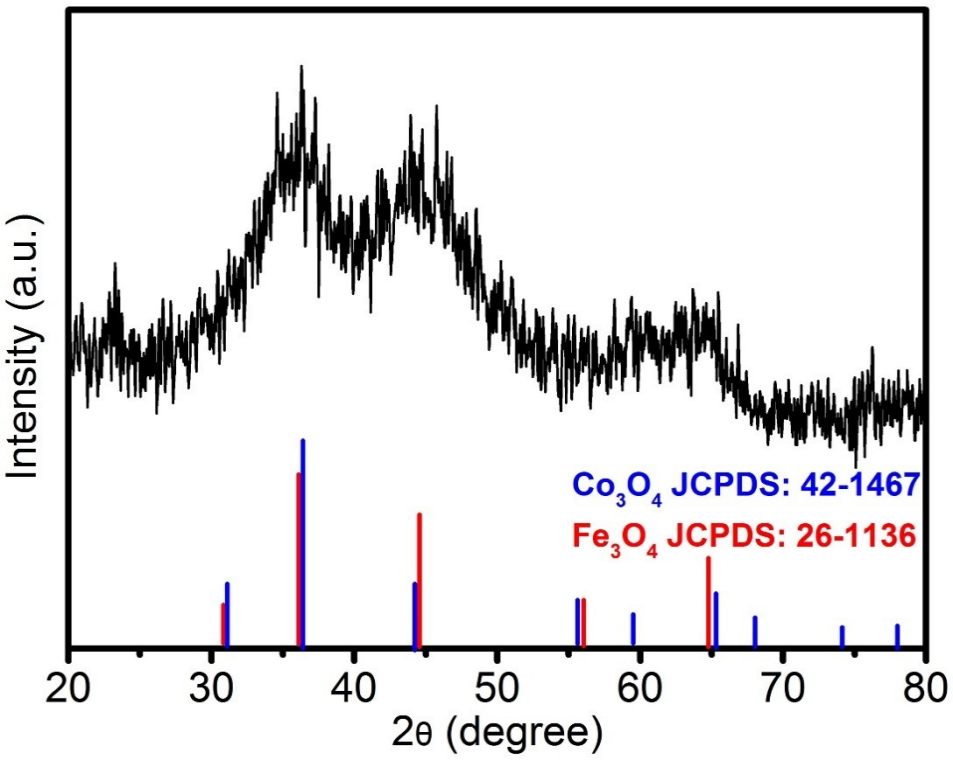

Figure S11. XRD spectrum of CoFeBO NS sample**.**

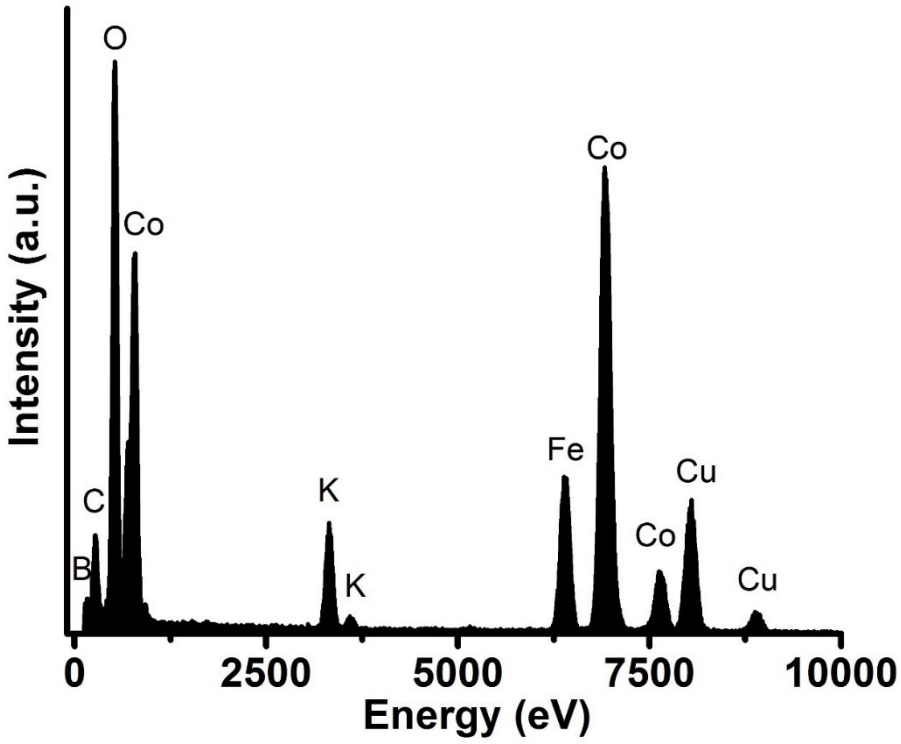

Figure S12. EDX spectrum of CoFeBO NS sample.

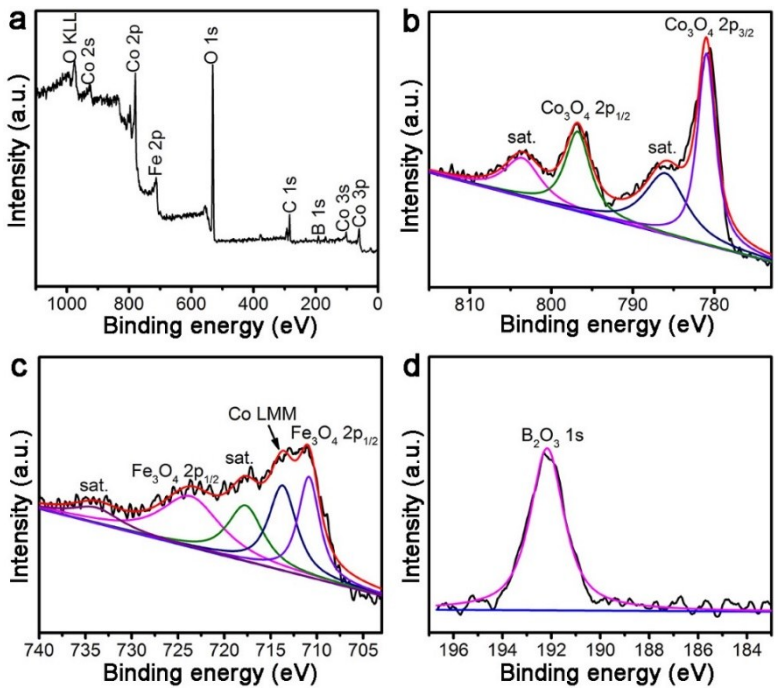

Figure S13. (a) XPS survey spectrum for CoFeBO NS; core level of XPS spectrum in the (b) Co 2p, (c) Fe 2p, and(d) B 1s for CoFeBO NS.

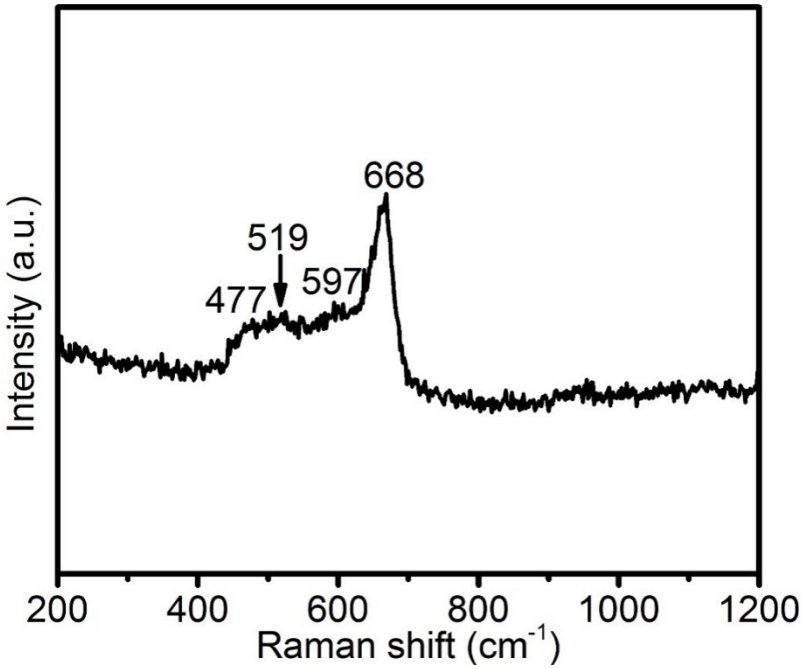

Figure S14. Raman spectrum of CoFeBO NS sample. The bands at 477, 519, 597 and 668 cm^{-1} are ascribed to the E_g, F_{2g}^1 and F_{2g}^2 and A_{1g} characteristic mode of Co_3O_4, respectively (J. Phys. C: Solid State Phys, **1988**, 21 L199-L201). It is also likely that some Fe_3O_4 contributes to the intensity of the 668 cm^{-1} band (Electrochimica acta, **1992**, 37: 2747-2754). These data suggests that the surface of the CoFeBO NS sample is covered by a layer of metal oxides, including Co_3O_4 and magnetite.

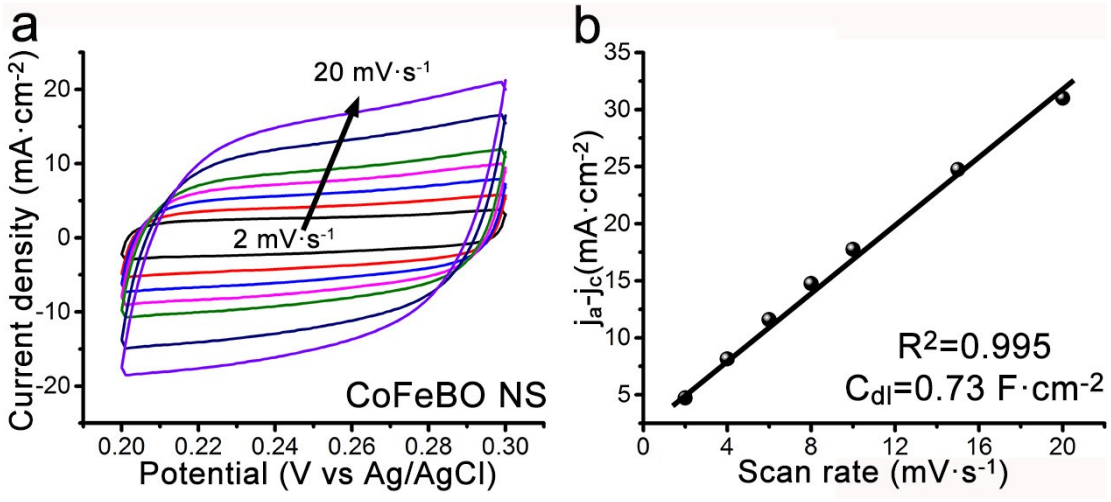

Figure S15. (a) Cyclic voltammograms recorded for the CoFeBO NS electrode at various scan rates in the non-Faradaic region in 1.0 M KOH. (b) Scan rate dependence of the current densities of the CoFeBO NS electrode at 0.25V vs Ag/AgCl . The slope of the lines in (b) is the effective electrochemically active surface area of CoFeBO electrodes.

Table S1. Comparison of Co NS and other non-noble metal electrocatalysts for HER in 1 M KOH.

Material	Overpotential η (V) @ corresponding j	Current density j(mA·cm^{-2})	Tafel slope (mV·dec^{-1})	Reference
MoB microparticles	~0.24	20	59	Angew. Chem., Int. Ed. 2012, 51, 12703
CoP nanowire arrays	0.209	10	129	J. Am. Chem. Soc. 2014, 136, 7587
Co-NRCNTs	0.37	10	69	Angew. Chem., Int. Ed. 2014, 53, 4372
MoP nanoparticles	~0.13	10	48	Energy Environ. Sci. 2014, 7, 2624
NiO/Ni@CNT	0.8	10	82	Nat. Commun. 2014 DOI:10.1038/ncomms5695
MoC_x nanooctahedrons	0.151	10	59	Nat. Commun. 2015 DOI:10.1038/ncomms7512
Mo_xC-Ni@NCV	0.126	10	93	J. Am. Chem. Soc. 2015, 137, 15753
CoO_x@CN	0.232	10	115	J. Am. Chem. Soc. 2015, 137, 2688
Ni-MoS_2	98	10	60	Energy Environ. Sci. 2016, 7, 2624
Co NS	**0.02**	**10**	**42.6**	**This work**
	0.06	**100**		
	0.083	**400**		

Table S2. Comparison of CoFeBO NS and other non-noble metal electrocatalysts for OER in 1.0 M KOH

Material	Overpotential η (V) @ corresponding j	Current density j ($mA \cdot cm^{-2}$)	Tafel slope ($mV \cdot dec^{-1}$)	Reference
Co_3O_4 NWs	0.42	13.1	72	Adv. Energy Mater. 2014, DOI: 10.1002/aenm.201400696
γ-CoOOH	0.3	10	38	Angew. Chem., Int. Ed. 2015, 54, 8722
Ni_2P	0.29	10	47	Energy Environ. Sci. 2015, 8, 2347
$Fe_{0.1}Ni_{0.9}O$	0.297	10	37	ACS nano, 2015, 9, 5180
NiFe/NF	0.27	80	28	Nat. Commun. 2015, 6, 6616
Co_4N	0.257	10	44	Angew. Chem., Int. Ed. 2015, 127,14923
Ni_3S_2/NF	0.26	10	——	J. Am. Chem. Soc. 2015, 137, 14023
CoP nanorods	0.29	10	65	Adv. Funct. Mater. 2015, 25, 7337
$CoSe_2$ sheets	0.47	73	64	Angew. Chem., Int. Ed. 2015, 54, 12004
Co-B_i NS/G	0.29	10	53	Angew. Chem., Int. Ed. 2016, 55, 2488
FeOOH/Co/FeOOH	~0.25	20	32	Angew. Chem., Int. Ed. 2016, 55, 3694
CoMnP	0.33	10	61	J. Am. Chem. Soc. 2016, 138, 4006
Co-P film	0.345	10	47	Angew. Chem., Int. Ed. 2015, 54,6251
	0.413	100		
	0.463	500		
h-NiS_x	0.18	10	96	Adv. Energy Mater., 2016, DOI: 10.1002/aenm.201502333
	0.217	100		
	0.316	500		
CoFeBO NS	**0.24**	**10**	**53**	**This work**
	0.28	**100**		
	0.31	**500**		

Table S3. Comparison of CoFeBO NS ‖ Co NS and other non-noble metal electrocatalysts for overall water splitting in 1.0 M KOH.

Material	Voltage (V)	Current density ($mA \cdot cm^{-2}$)	Reference
NiSe NWs ‖ NiSe NWs	1.63	10	Angew. Chem., Int. Ed. 2015, 54, 9351
Ni_5P_4 ‖ Ni_5P_4	~1.7	10	Angew. Chem., Int. Ed. 2015, 127, 12538
CoP ‖ CoP	1.74	100	Angew. Chem., Int. Ed. 2015, 54, 6251
CoP-MNA ‖ CoP-MNA	1.62	10	Adv. Funct. Mater. 2015, 25, 7337
Ni/NiP ‖ Ni/NiP	1.61	10	Adv. Funct. Mater. 2016, DOI: 10.1002/adfm.201505626
NiFe LDH ‖ NiFe LDH	1.7	10	Science, 2014, 345, 1593
	1.8	20	
CoMnO@CN ‖ CoMnO@CN	1.7	54	J. Am. Chem. Soc., 2015, 137, 14305
	1.8	108	
$NiFeO_x$ ‖ $NiFeO_x$	1.7	30	Nat. Commun. 2015, DOI:10.1038/ncomms8261
	1.8	100	
EG/$Co_{0.85}Se$/NiFe-LDH ‖ EG/$Co_{0.85}Se$/NiFe-LDH	1.67	10	Energy Environ. Sci., 2016, 9, 478
	1.71	20	
$NiCo_2O_4$ ‖ $Ni_{0.33}Co_{0.67}S_2$	1.72	10	Adv. Energy Mater. 2015, DOI: 10.1002/aenm.201402031.
NiFe LDH ‖ NiO/Ni-CNT	1.5	10	Nat. Commun. 2014, DOI:10.1038/ncomms5695.
NiFe LDH ‖ Cr_2O_3/NiO-Ni	1.75	200	Angew. Chem., Int. Ed. 2015, 54, 11989
CoFeBO NS ‖ Co NS	**1.5**	**10**	**This work**
	1.62	**100**	
	1.68	**200**	